P02 Development of a method to measure trace level of uranium and thorium in scintillation films

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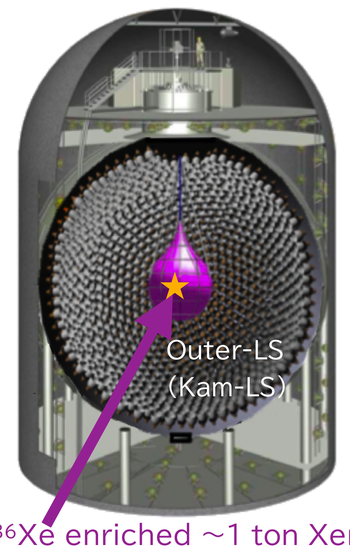
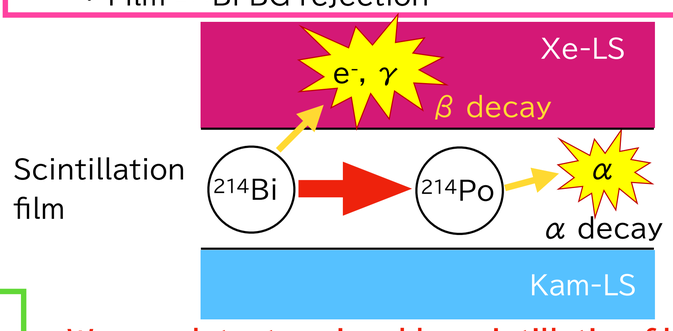
1. KamLAND2-Zen experiment



・^{136}Xe enriched ~1 ton Xenon in Xe-loaded LS (Xe-LS)

Goal : covering the IO region
Target sensitivity :
$T_{1/2}$: 2×10^{27} years
$\langle m_{\beta\beta} \rangle$: 20 meV in 10 years

Detector upgrade :
× ~5 increase effective light yield
(×1.8 light correction w/ Winstone cones)
(×1.4 High light yield LS based on LAB)
(×1.9 High QE 20-inch PMTs)
state-of-the-art electronics : MoGURA2
☆Scintillation inner-balloon film (this poster)
→ Film ^{214}Bi BG rejection



We can detect α signal by scintillation film

Understanding RIs in the scintillation film & wavelength shifter is necessary

2. Motivation

- **Material screening for detector components (organic materials) with high sensitivity**
- KamLAND2-Zen's requirements
  - Scintillation film (PEN film)
    ➡ ∅ (1) ppt* for ^{238}U and ^{232}Th
  - Bis-MSB (Wavelength shifter) :
    ➡30 ppt for ^{238}U, 100 ppt for ^{232}Th

★ **As a first step, establish a method to measure ppt level ^{238}U and ^{232}Th in organic materials**


Polyethylene naphthalate (PEN) film


Bis-MSB Wavelength Shifter

✓ Careful sample & labware treatment
✓ Procedure blank, Recovery measurement
• Next, establish a purification method (Bis-MSB)
➡ Eventually, sub-ppt or ppq level measurement

M. Kurasawa's master thesis (2023) and my work

Collaborative research of RCNS and CRiES

* ppt = 10^{-12} g/g_{sample}

3 Methods of Measuring Radioactive Elements

Sample preparation with special care in clean room


PEN film


Bis-MSB

beakers and Polyethylene (PE) bags : soaked in HNO_3 (TAMAPURE AA-100) for several days
0.7 mol/L for beakers, 0.15 mol/L for PE bags

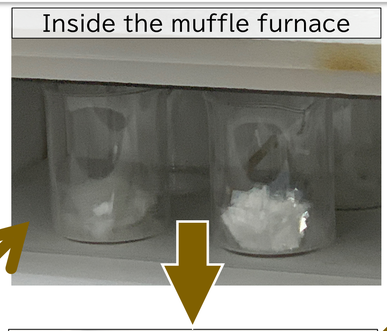
①Cleaning samples


Ultrasonic cleaning w/ ultrapure water

②Ashing

PYRO microwave ashing system

Inside the muffle furnace



Sample after ashing

③Dissolving


5 ml (~5 $g_{solution}$) diluted TAMAPURE AA-100 HNO_3

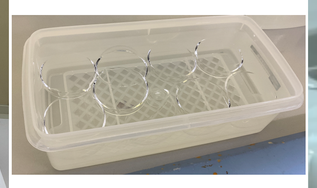

④ICP-MS measurement
Agilent 8800

- Special tune for ^{238}U, ^{232}Th measurement
  - (nebulizer, lens etc.)
- Detection limit of the instrument
  - from ultrapure-water sample
    ➡ 1.1 ppq** (^{238}U), 8.1 ppq (^{232}Th)
- Further improvement :Aridus3
  - solvent removal module
  - expected ×5 improvement

**ppq = fg/$g_{solution}$

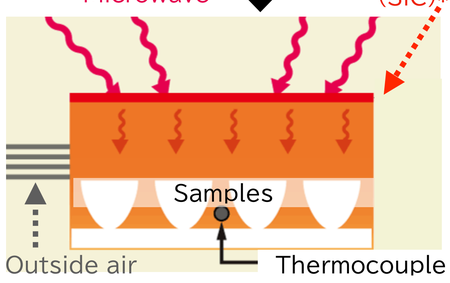
①' Careful cleaning of labware to reduce contamination at Tsukuba


①soaked in 1.5 mol/L EL-grade HNO_3 @ 90℃, > 2 d
②soaked in ultrapure water (90℃, > 2 d)


③soaked in TMSC solution until just before use (TMSC : high-purity alkaline cleaning solution)

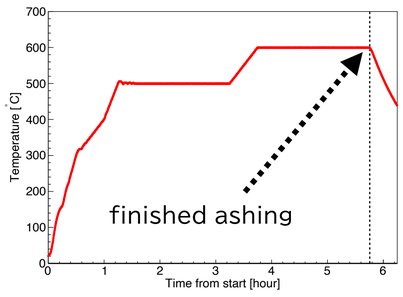

④flushed w/ ultrapure water
⑤rinsed 1 mL 15.2 mol/L HNO_3 (TAMAPURE AA-100)
⑥cleaned 2 mL 0.15 mol/L HNO_3 ×2 times

Heating element (SiC)*



* : Absorbs microwaves to heat the inside of the furnace

Temp. variation during ashing



~2g PEN film can be ashed about 6hr operation + ~2 hr cooling time

4-1 Result (Procedure blank)

- Check the detection limit of the procedure (MDL) due to contamination from washing, ashing, and dissolution process

・washing
・ashing process
・making solution
→ ICP-MS

- MDL in this poster is set as : Average + 3× Std. Dev.
- Confirm a few ppt level sensitivity

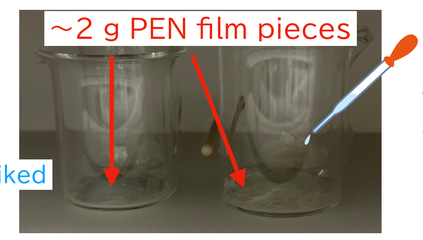
(Current case : 1 pg/$g_{solution}$ ~ 2.5 pg/g_{sample})

Blank#	^{238}U	^{232}Th
1	0.19	1.00
2	0.06	0.49
3	0.14	0.43
4	0.12	0.34
5	0.23	0.30
6	0.26	0.29
7	0.15	0.29
8	0.60	0.55
9	0.12	0.24
Average	0.21	0.44
Std. Dev.	0.16	0.23
MDL	0.69	1.13

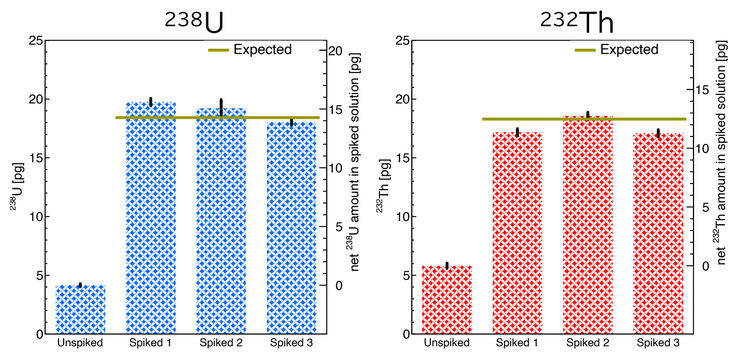
unit : pg/$g_{solution}$

4-2 Result (Addition recovery experiment)

- Check how much ^{238}U and ^{232}Th could be collected from sample using this method
- spiked a known amount of ^{238}U and ^{232}Th to the sample
  ➡ ashing, dissolution, ICP-MS



spiked
diluted XSTC-331
3 samples

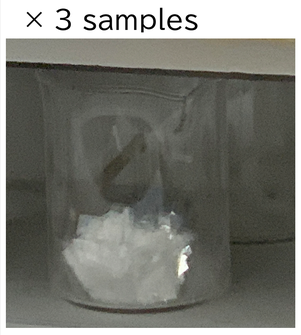


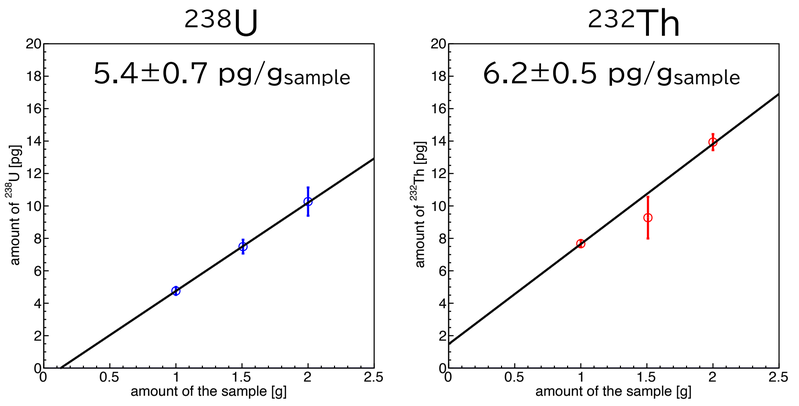
Expected (^{238}U, ^{232}Th) = (14.3±0.2, 12.5±0.4) [pg]
Observed = (14.6±0.5, 11.8±0.5) [pg]
Recovery Rate = 102.4±3.6% (^{238}U)
= 94.3±3.8% (^{232}Th) : ~100%

- This method can collect trace amount of ^{238}U and ^{232}Th without loss

4-3 Result (PEN film measurement)

3 weights (1.0, 1.5, 2.0 g) × 3 samples





slope : concentration of ^{238}U, ^{232}Th in the sample*

✓ **Demonstrate O(ppt) ^{238}U, ^{232}Th measurement in organic materials**
✓ This result meets KamLAND2-Zen's requirement

*Y. Takaku et al, Journal of Analytical Atomic Spectrometry 9, 1385 (1994)

5 Summary and future improvement

✓ Detection limit of method : a few ppt level both ^{238}U and ^{232}Th
✓ Almost 100% recovery rate (collection efficiency)
✓ Demonstrate U, Th measurement in PEN film (organic material)
★ **Submitted to PTEP**
➡ Toward sub-ppt level measurement :
- improving environment for pretreatment
  (more HEPA filter, radio-pure synthetic quartz, clean fume hood etc.)

▸ In parallel, check PEN characteristic
(QE, Light yield, transparency etc.)

* : presenter

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